Field-induced coupled superconductivity and spin density wave order in the Heavy Fermion compound $CeCoIn_5$

J. Spehling,[1] R. H. Heffner,[2] J. E. Sonier,[3] N. Curro,[4] C. H. Wang,[2,5] B. Hitti,[6] G. Morris,[6] E. D. Bauer,[2] J. L. Sarrao,[2] F. J. Litterst,[7] and H.-H. Klauss[1,*]

[1]*Institut für Festkörperphysik, TU Dresden, D-01069 Dresden, Germany*
[2]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
[3]*Department of Physics, Simon Fraser University, Burnaby, Canada*
[4]*Departments of Physics, University of California, Davis, USA*
[5]*University of California, Irvine, California 92697, USA*
[6]*Tri-Meson Facility, University of British Columbia, Vancouver, Canada*
[7]*Institut für Physik der Kondensierten Materie, TU Braunschweig, D-38106, Germany*

(Dated: October 2, 2009)

The high field superconducting state in $CeCoIn_5$ has been studied by transverse field muon spin rotation measurements with an applied field parallel to the crystallographic c-axis close to the upper critical field $\mu_0 H_{c2} = 4.97$ T. At magnetic fields $\mu_0 H \geq 4.8$ T the muon Knight shift is enhanced and the superconducting transition changes from second order towards first order as predicted for Pauli-limited superconductors. The field and temperature dependence of the transverse muon spin relaxation rate σ reveal paramagnetic spin fluctuations in the field regime from 2 T$\leq \mu_0 H < 4.8$ T. In the normal state close to H_{c2} correlated spin fluctuations as described by the self consistent renormalization theory are observed. The results support the formation of a mode-coupled superconducting and antiferromagnetically ordered phase in $CeCoIn_5$ for H directed parallel to the c-axis.

PACS numbers: 74.70.Tx, 76.75.+i, 74.25.Ha

In a conventional type-II superconductor the magnetic upper critical field H_{c2} is determined by the orbital effect, i.e. the formation of vortices due to orbital screening currents which increase the kinetic energy of the Cooper pairs. If the material is layered or if the effective mass of the quasiparticles becomes large the orbital effect is reduced and superconductivity can be found up to higher applied magnetic fields. In this case the so-called Pauli limit for spin-singlet superconductivity is reached when the magnetic Zeeman energy, $\mu_B H$, overcomes the binding energy of the Cooper pairs [1]. Close to the Pauli limit, complex quantum ground states like the Fulde-Ferrel-Larkin-Ovshinnikov (FFLO) superconducting (SC) phase [2] and other modulated sc phases with mixed singlet-triplet order (pair density wave, PDW) coupled to a spin density wave (SDW) magnetic order [3, 4, 5], are proposed. $CeCoIn_5$, a layered heavy fermion superconductor with a critical temperature $T_c = 2.3$ K, is a model system to test these predictions.

The SC state in $CeCoIn_5$ in zero or low magnetic fields is established to be of spin-singlet $d_{x^2-y^2}$ symmetry [6]. In high magnetic fields close to H_{c2}, the SC-to-normal phase transition becomes first order at a temperature T_0 [7, 8]. Moreover, a number of experiments provide evidence for a second order phase transition inside the SC state near H_{c2} for fields applied along the a-direction [7, 9, 10, 11]. This high-field SC (HFSC) state was initially considered as the realization of the FFLO state [2]. The FFLO state is characterized by a spatial modulation of the SC order parameter along the field direction perpendicular to the vortex lines [13], resulting in a periodic array of planes of normal paramagnetic electrons. However, NMR experiments in the HFSC state of $CeCoIn_5$ for $H\|$a revealed a site-dependent line broadening at inequivalent In positions upon entering the low-temperature SC phase from the homogenous SC phase. This was interpreted as evidence for antiferromagnetic (AF) order [14]. Subsequent neutron diffraction experiments with fields applied along (1,-1,0) found indeed an incommensurate SDW at $\mathbf{Q} = (0.44, 0.44, 0.5)$ in the HFSC region with a small Ce magnetic moment $\sim 0.15\mu_B$ [3]. SDW order and superconductivity simultaneously disappear at the same upper critical field, indicating a coupling of the SC and AF order parameters. In this spatially inhomogeneous superconducting state the SC condensate carries a finite momentum corresponding to the modulation wave vector of the magnetic order q. The difference between the order parameter in such a 'Q-phase' and the FFLO order parameter is that in the 'Q-phase' q is H independent and of short wave length, and also that mixed singlet and triplet SC order parameters can occur.

For the field direction $H\|$c the existence of such phases in $CeCoIn_5$ is still under debate and remains a matter of great interest [7, 15, 16]. In a local probe experiment like NMR or μSR, a long-wave-length modulation of the SC condensate density is expected to cause an additional resonance line due to the introduced paramagnetic planes [17]; therefore such experiments are ideally suited to identify the FFLO phase. NMR measurements by Kumagai et al. revealed such a double peak structure only close to T_c [15]. However, this structure may be

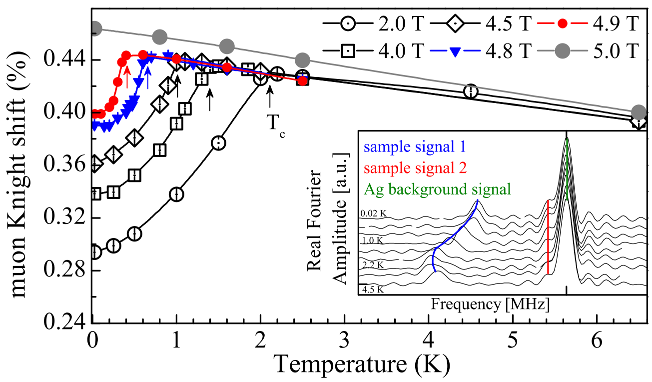

FIG. 1: (color online). Muon Knight shift as a function of temperature at various magnetic fields. Arrows indicate the SC transition temperatures T_c. Lines are guides to the eye. Inset: Fourier transformed muon spin precession spectra at different temperatures at an applied field of 2 T.

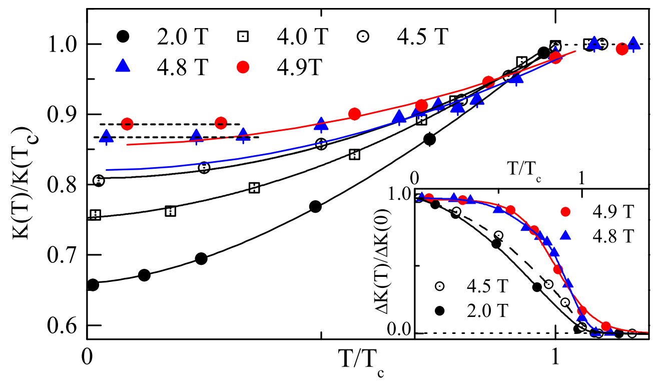

FIG. 2: (color online). Normalized muon Knight shift $K(T)/K(T_c)$ in the SC state as a function of the reduced temperature. Solid lines denote a quadratic T-dependence. Inset: Normalized muon Knight shift $\Delta K(T)/\Delta K(T \to 0)$ as a function of T/T_c.

caused by RF heating, as demonstrated by Mitrovic et al. [18].

In this Letter, we present a detailed transverse field (TF) muon spin rotation (μSR) study on single crystalline $CeCoIn_5$. We focus on the low-temperature (0.02 to 7 K) and high-field (2 to 5 T) part of the $H-T$-phase diagram for $H \| c$. Our experiments reveal a field-induced enhanced local spin susceptibility close to H_{c2}, which becomes temperature-independent at low temperatures, consistent with the appearance of a triplet component in the SC order parameter. Our results further show a strong nonlinear enhancement of the static line width and the dynamic $(T_1T)^{-1}$ relaxation rate with increasing field below H_{c2} consistent with a coupled SC and AF order. Our data do not show the appearance of an additional peak at the paramagnetic position in the local probe frequency spectrum, as predicted for a FFLO state with a long wave length modulation q.

Single crystals of $CeCoIn_5$ were grown in In flux [19]. The measurements were performed at the M15 beam line, TRIUMF, Vancouver, Canada, using a top loading dilution refrigerator. External magnetic fields up to 5 T were applied parallel to the crystallographic c-axis transverse to the initial μ-spin polarization. All experiments were performed under field cooled conditions. The muon Knight shift $K = \frac{\omega_\mu - \omega_{Ag}}{\omega_{Ag}}$ can be expressed as $K = (A_c + A_{dip}) \cdot \chi_{4f} + K_{dem} + K_0 + K_{dia}$. Here, $A_c + A_{dip}$ describes the contact and dipolar coupling of the muon to the $4f$ susceptibility χ_{4f}, K_{dem} is the correction for demagnetization and Lorentz fields, K_0 is the isotropic temperature-independent contribution from non-$4f$ conduction electrons and K_{dia} is due to flux expulsion in the SC state. In type-II superconductors the last term can lead to an internal field distribution $p(B_{local})$, with $\omega_\mu = \gamma_\mu B_{local}$ and $\gamma_\mu/2\pi = 135.5342$ MHz/T, which can be measured via a contribution to the static line width σ_s.

The Fourier transformed muon spin precession spectra (inset, fig. 1) show one distinct symmetric line (sample signal 1) and one non-symmetric line which is due to the overlap of sample signal 2 and a temperature independent Ag background signal. Two sample signals, distinct by their different precession frequencies, indicate two magnetically inequivalent muon sites. Calculations of the dipolar hyperfine coupling tensor A_{dip} for localized Ce-$4f$ moments interacting with the muon spin identify signal 2 with the muon stopping site $(\frac{1}{2},\frac{1}{2},\frac{1}{2})$ and signal 1 with the muon stopping sites $(\frac{1}{2},0,0)$ and $(0,\frac{1}{2},0)$. The hyperfine coupling at site 2 is an order of magnitude smaller than at site 1. Hence, it strongly overlaps with the Ag signal and, therefore, our discussion will focus solely on signal 1 due to its clear separation. Moreover, sample signal 2 shows nearly no frequency shift with respect to the Ag background signal at all temperatures studied. Since any demagnetization or Lorentz fields as well as the diamagnetic orbital contribution in the SC state to $K(T)$ would be the same at all muon sites we conclude that those contributions are negligible and temperature independent. Therefore, $K(T)$ directly samples the local spin susceptibility $\chi_{4f}(T)$.

In the SC phase at all examined field strengths signal 1 is homogeneous and does not show a splitting with an additional line at the paramagnetic position as expected for a FFLO phase [17]. The main panel of fig. 1 shows the muon Knight shift $K(T)$ as a function of temperature for various applied fields. As the temperature is lowered towards T_c, $K(T)$ slightly increases due to a Curie behavior of the spin polarization. Below T_c, $K(T)$ reflects the suppression of $\chi_{4f}(T)$ due to spin singlet pairing. In the case of s-wave spin singlet pairing, $\chi_{4f}(T)$ is expected to vanish for $T \to 0$ K [20]. However, spin-orbit (SO) scattering by impurities and unconventional pairing states,

e.g. d-wave pairing with nodes in the gap function or spin-triplet pairing can cause $\chi_{4f}(0)$ to be finite and to increase toward its normal state value. Increasing the external field suppresses T_c towards smaller temperatures. At an applied of 5 T, superconductivity is completely suppressed. An upper critical field of $\mu_0 H_{c2} = 4.97$ T is deduced from a fit to the $H(T_c)$ data (not shown). Note, the small 4% increase of the Knight shift in 5 T external field indicates a slightly enhanced spin polarization in the paramagnetic state.

The inset of fig. 2 depicts the Knight shift change $\Delta K(T)$ normalized to $\Delta K(T \to 0)$ as function of the reduced temperature. Whereas a relatively small change occurs from 2 T to 4.5 T, we observe a substantial change between 4.5 T to 4.8 T indicating a field-induced change from a second order towards a first order phase transition between the normal and the SC state. Note that SC fluctuations [12] may modify the first order transition into a nearly continuous crossover. The change from second towards first order appears at a corresponding temperature of $T_0 \approx 0.65$ K $\approx 0.3 T_c(H = 0)$. This agrees well with reported values [7, 8] and the predictions by Gruenberg and Gunther for Pauli-limited type-II-superconductors [21].

The T-dependence of the local magnetization in the SC state is expected to be quadratic for high magnetic fields when $\mu_B B > k_B T$ [22, 23], with a residual value χ_{4f} [24]. The main panel of fig. 2 displays $K(T)$ normalized to $K(T_c)$ in the SC state. For $\mu_0 H < 4.8$ T the data agree well with the predicted quadratic T-dependence. For $\mu_0 H \geq 4.8$ T we observe a discrepancy. This discrepancy close to H_{c2} is particularly pronounced at very low temperatures where the local spin susceptibility is enhanced and nearly temperature independent (dashed lines) in comparison to the $\mu_0 H < 4.8$ T data. This result is consistent with Knight shift calculations from Aperis et al. considering a field-induced coexistence of d-wave spin-singlet superconductivity with staggered π-triplet superconductivity [5, 25, 26, 27]. Moreover, our Knight shift data mirror NMR results showing a comparable behavior of the spin susceptibility for fields along a-axis [28] indicating a common high field ground state in $CeCoIn_5$ regardless of the direction of the external field.

Fig. 3 shows the temperature dependence of the muon spin relaxation rate σ at various applied magnetic fields. For all field strengths ($\mu_0 H \geq 2T$) we find a *reduction* of σ in the SC state with decreasing temperature. This is contrary to the expected increase of the static line width σ_s in the vortex state due to the formation of the flux line lattice (FLL) and the decrease of the in-plane magnetic penetration depth λ_{ab} in the SC phase, i.e. $\sigma_s \propto 1/\lambda_{ab}^2$. Note that our results differ considerably from low-field (0.3 T) TF-μSR data, which show a slight *increase* of σ in the SC state [29].

Since there is no common mechanism which leads to a reduction of the static line width below a conventional SC phase transition we now consider that the transverse field muon relaxation rate σ is the sum of a static line width σ_s and a dynamic $1/T_1$ contribution. To extract $1/T_1$ we assume a temperature-independent σ_s [36]. This is reasonable since at 5 T external field ($B > B_{c2}$) there is nearly no change in the relaxation rate with temperature in the T-regime studied. A sizeable dynamic contribution is deduced only for those field values at which a SC transition is found. Therefore, at 5 T external field σ is decreased in comparison with the data at lower field values in the paramagnetic regime.

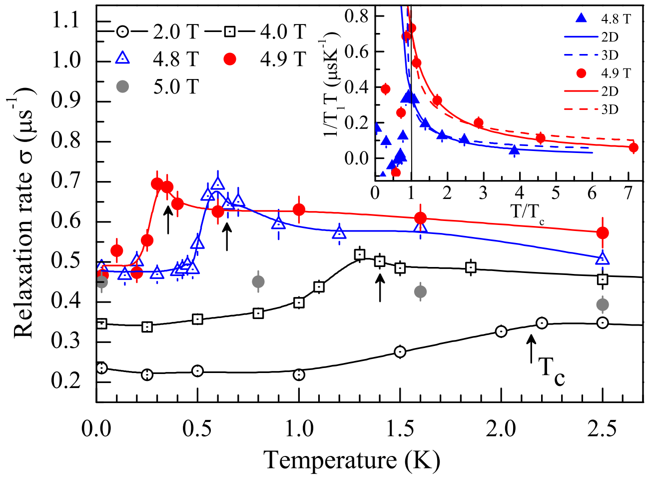

FIG. 3: (color online). Muon relaxation rate σ as a function of temperature at various magnetic fields. Arrows indicate T_c derived from Knight shift data. Lines are guides to the eye. Inset: $\frac{1}{T_1 T}$ vs. T/T_c at 4.8 T and 4.9 T. Solid lines denote fit results according to Moriya's SCR theory. See text for details.

The dynamic muon relaxation rate $\frac{1}{T_1}$ measures the magnetic fluctuation spectrum of the electronic spin system at frequency ω_μ, i.e., $\frac{1}{T_1 T}$ is proportional to the $\mathbf{q}$-integrated dynamic magnetic susceptibility $\chi''(\omega_\mu)$. We plot the extracted dynamic muon relaxation depicted as $\frac{1}{T_1 T}$ in the inset of fig. 3 for applied fields of 4.8 T and 4.9 T and for $T > T_c$. For a metal one expects a Korringa behavior, i.e. $\frac{1}{T_1 T}$ to be constant. Instead, we observe that the T-dependence of $\frac{1}{T_1 T}$ follows a power-law,

$$\frac{1}{T_1 T} \propto (T - T_N)^{-\beta}, \qquad T > T_c, \tag{1}$$

diverging at finite temperatures below T_c. This indicates a critical slowing down of magnetic spin fluctuations close to a second order magnetic phase transition below T_c. The data can be described equally well with a critical dynamic exponent $\beta = 1$ for 2-D or $\beta = 0.5$ for 3-D AF correlations according to Moriya's self consistent renormalization (SCR) theory [30]. These magnetic correlations induce the dynamic muon spin relaxation dominating the muon relaxation rate σ for $T > T_c$. Note, at all measured field strengths below 5 T, $\frac{1}{T_1 T}$ can be described by a power-law indicating the presence of critical

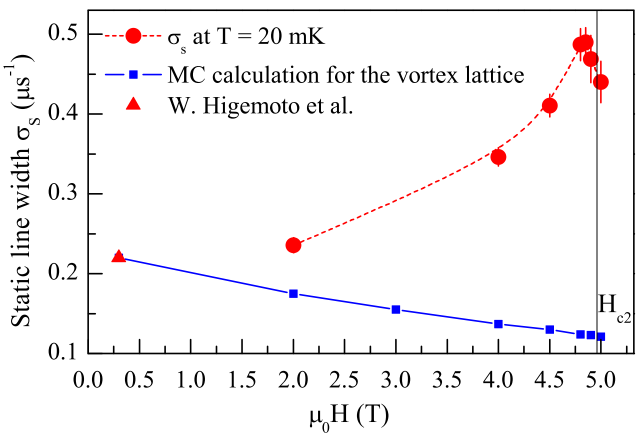

FIG. 4: (color online). Field dependence of the static line width σ_s at $T = 20$ mK (red circles). The blue squares depict calculated values for an Abrikosov vortex lattice with $\lambda = 5500$Å and $\xi = 46.84$Å. Lines are guides to the eye.

spin fluctuations. Whereas our μSR data at 2 T and 4 T diverge at $T = 0$ K indicating an AF quantum phase transition [31], the high field data show a divergence at finite temperatures below T_c ($T_N/T_c \approx 0.5$ assuming 2-D and $T_N/T_c \approx 0.9$ assuming 3-D correlations) as if there were an accompanying magnetic phase transition. In the SC state due to spin singlet formation a reduction of $\frac{1}{T_1T}$ is observed. Our relaxation rate data agree qualitatively well with the calculated spin-lattice relaxation rate $\frac{1}{T_1T}$ in Ref. [5]. This further supports the suggested scenario of SDW order triggered by the emergence of a π-triplet pairing component in the superfluid [4, 5, 25]. In contrast to Ref. [5] in the normal state we observe an increase of $1/T_1T$ as T_C is approached from higher temperatures which is attributed to the vicinity to a magnetic phase transition.

The field dependence of the static muon relaxation rate σ_s at $T = 20$ mK is depicted in fig. 4. It shows a non-linear increase of the internal field distribution in the SC state. This is in conflict with the Ginzburg-Landau paradigm, which predicts a monotonically decrease of σ_s for $H > H_{c1}$, with increasing field as verified by Monte Carlo calculations [37]. The low-field data point [29] fits well into this expected field dependence as denoted by the red triangle. The strongly enhanced static line width around 4.85 T is associated with a field-induced magnetic order inside the SC phase. Our findings are similar to recent neutron diffraction data for $H\|c$, which report an unusual enhancement of the vortex lattice form factor consistent with localized spins inside the vortex cores [16]. The decrease of the static line width at 5 T external field implies that the additional source of magnetism vanishes simultaneously at the same critical field H_{c2}. This confirms the intimate coupling between magnetism and superconductivity [3, 4, 5] in $CeCoIn_5$.

In conclusion, we performed transverse field μSR measurements at high magnetic fields up to 5 T with $H\|[001]$ to examine the microscopic nature of the SC state in $CeCoIn_5$. The local spin susceptibility $\chi_{4f}(T) \propto K(T)$ is reduced in the SC state consistent with spin-singlet superconductivity. Close to H_{c2} we find an unusual enhanced and temperature independent spin susceptibility at low temperatures consistent with theoretical calculations [5] of the appearance of a staggered spin-triplet SC component coexisting with the $d-$wave singlet superconductivity. We further observed a strong non-linear increase of the static line width as well as the dynamic $\frac{1}{T_1T}$ relaxation rate with increasing field strength in the SC state. These results support a field-induced coupled SC and AF phase transition as described by a pair density wave (PDW) theory discussed e.g. by Agterberg et al. and Aperis et al. [4, 5].

This work was performed at the Tri-University Meson Facility (TRIUMF), Vancouver, Canada. We acknowledge with thanks the help of D. Arsenau, S. Kreitzman and the TRIUMF accelerator crew. Work at TU Dresden is supported by the DFG under Grant No. KL1086/8-1. Work at UC Irvine is supported by U.S. DOE Grant No. DE-FG02-03ER46036. Work at Los Alamos was performed under the auspices of the U. S. DOE/Office of Science and supported by the Los Alamos LDRD program.

* Electronic address: h.klauss@physik.tu-dresden.de

[1] A. M. Clogston, Phys. Rev. Lett. **9**, 266 (1962).
[2] P. Fulde and R. A. Ferrell, Phys. Rev. **135**, A550 (1964); A. I. Larkin and Yu. N. Ovchinnikov, Zh. Eksp. Teor. Fiz. **47**, 1136 (1964).
[3] M. Kenzelmann *et al.*, Science **321**, 1652 (2008).
[4] D. F. Agterberg, M. Sigrist and H. Tsunetsugu, Phys. Rev. Lett. **102**, 207004 (2009).
[5] A. Aperis, G. Varelogiannis and P. B. Littlewood, arXiv:0902.0553 (unpublished).
[6] K. Izawa *et al.*, Phys. Rev. Lett. **87**, 057002 (2001).
[7] A. Bianchi *et al.*, Phys. Rev. Lett. **91**, 187004 (2003).
[8] T. Tayama *et al.*, Phys. Rev. B **65** 180504 (2002).
[9] A. Radovan *et al.*, Nature **425**, 51 (2003).
[10] C. Capan *et al.*, Phys. Rev. B **70**, 134513 (2004).
[11] K. Kakuyanagi, *et al.*, Phys. Rev. Lett. **94**, 047602 (2005).
[12] H. Adachi *et al.*, cond-mat/0303540.
[13] Y. Matsuda and H. Shimahara, J. Phys. Soc. Jpn. **76**, 051005 (2007), and references therein.
[14] B. L. Young *et al.*, Phys. Rev. Lett. **98**, 036402 (2007).
[15] K. Kumagai *et al.*, Phys. Rev. Lett. **97**, 227002 (2006).
[16] A. Bianchi *et al.*, Science **319**, 177 (2008).
[17] M. Ichioka, K. Machida, Phys. Rev. B **76**, 064502 (2007).
[18] V. F. Mitrovic *et al.*, arXiv: 0806.3600 (2008).
[19] C. Petrovic *et al.*, J. Phys. Cond. Mat. **13**, L337-L342 (2001).
[20] K. Yosida, Phys. Rev. **110**, 769 (1958).
[21] L. W. Gruenberg and L. Gunther, Phys. Rev. Lett. **16**,

996 (1966).

[22] A. B. Vorontsov and M. J. Graf, cond-mat/0507479.

[23] K. Yang and S. L. Sondhi, Phys. Rev. B **57**, 8566 (1998).

[24] Note, the results in Ref. [22] refer to calculations with $H\|$ab-plane.

[25] A. Aperis *et al.*, J. Phys. Condens. Matter **20**, 434235 (2008).

[26] G. C. Psaltakis, G.C. and E. W. Fenton, J. Phys. C **16**, 3913-3932 (1983).

[27] B. Kyung, Phys. Rev. B **62**, 9083 (2000).

[28] V. F. Mitrovic *et al.*, Phys. Rev. Lett. **97**, 117002 (2006).

[29] W. Higemoto *et al.*, J. Phys. Soc. Jpn. **71**, pp.1023-1026 (2001).

[30] T. Moriya and K. Ueda, Adv. Phys. **49**, 555 (2000).

[31] N. J. Curro *et al.*, Nature **434** (2005).

[32] E. H. Brandt, Phys. Rev. Lett. **78**, 11 (1997).

[33] M. A. Tanatar *et al.*, Phys. Rev. Lett. **95**, 067002 (2005) and references therein.

[34] P. M. C. Rourke *et al.*, Phys. Rev. Lett. **94**, 107005 (2005) and references within.

[35] D. F. Agterberg, T. M. Rice, and M. Sigrist, Phys. Rev. Lett. **78**, 17 (1997).

[36] $\sigma(20mK)$ is taken as the static line width σ_s since we can expect $\frac{1}{T_1} = 0$ at 0 K. To deduce σ_s in the normal state for fields $\mu_0 H > 4.8$ T we take the linear interpolated value between σ_s (2 T) and σ_s (5 T) since the unusual enhancement of the static line width around 4.85 T shown in fig. 4 is a unique feature of the combined superconducting/magnetic state. Therefore one can assume this enhancement is not present in the normal state at these magnetic field strengths.

[37] MC calculation of the internal field distribution due to the vortex lattice formation assuming a Cooper pair coherence length $\xi = 46.84$Å and a London penetration depth $\lambda = 5500$Å [29].